

Form PTO-1390 (Rev. 5-93) <b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE  ATTORNEY'S DOCKET NO. <b>H 2938 PCT/US</b>
		U.S. APPLICATION NO. (if known sec. 17 CER 1.5) <b>09/486677</b>
INTERNATIONAL APPLICATION NO. <b>PCT/EP98/05355</b>	INTERNATIONAL FILING DATE <b>August 24, 1998</b>	PRIORITY DATE CLAIMED <b>September 1, 1997</b>
<b>TITLE OF INVENTION</b> <b>COLD-STABLE FATTY ALCOHOL ALKOXYLATES</b>		
<b>APPLICANT(S) FOR DO/EO/US</b> <b>Ansgar Behler and Horst-Dieter Schares</b>		
<p>Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:</p> <ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)).             <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li> <li>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))             <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input checked="" type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). <b>(UNEXECUTED)</b></li> <li>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol> <p><b>Items 11. to 16. below concern other document(s) or information included:</b></p> <ol style="list-style-type: none"> <li>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>13. <input type="checkbox"/> A <b>FIRST</b> preliminary amendment             <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li>14. <input type="checkbox"/> A substitute specification.</li> <li>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>16. <input type="checkbox"/> Other items or information.:</li> </ol>		
<b>"Express Mail Post Office to Addressee" service Mailing Label Number</b> <b><u>EL541611884US</u></b>		

U.S. Application No. (if known see CFR 1.30) <b>09/486677</b>	INTERNATIONAL APPLICATION NO. <b>PCT/EP98/05355</b>	ATTORNEY'S DOCKET NUMBER <b>H 2938 PCT/US</b>	
<p>17. ■ The following fees are submitted:</p> <p><b>Basic National Fee (37 CFR 1.492(a)(1)-(5)):</b></p> <p>Search Report has been prepared by the EPO or JPO..... \$840.00</p> <p>International preliminary examination fee paid to USPTO (37CFR 1.482)..... \$670.00</p> <p>No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37CFR 1.445(a)(2))..... \$690.00</p> <p>Neither international preliminary examination fee (37CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$970.00</p> <p>International preliminary examination fee paid to USPTO (37CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... \$96.00</p>		CALCULATIONS      PTO USE ONLY	
<b>ENTER APPROPRIATE BASIC FEE AMOUNT</b> =		\$ 840 00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).		\$ 0 00	
Claims	Number filed	Number Extra	Rate
Total Claims	9 - 20 =	0	0 X \$18.00
Independent Claims	1 - 3 =	0	0 X \$78.00
Multiple dependent claims (s)(if applicable)		0	+ \$260.00
<b>TOTAL OF ABOVE CALCULATIONS</b> =		\$ 840 00	
Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).		\$ 0 00	
<b>SUBTOTAL</b> =		\$ 840 00	
Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37CFR 1.492(f)).		\$ 0 00	
<b>TOTAL NATIONAL FEE</b> =		\$ 840 00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		\$ 0 00	
<b>TOTAL FEES ENCLOSED</b> =		\$ 840 00	
		Amount to be: refunded	
		charged	
		\$840.00	
<p>a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.</p> <p>b. ■ Please charge my Deposit Account No. <u>50-1177</u> in the amount of <u>\$840.00</u> to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>00-0090</u>.</p> <p>c. ■ The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>50-1177</u>. A triplicate copy of this sheet is enclosed.</p> <p><b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.</b></p>			
<p>SEND ALL CORRESPONDENCE TO: Cognis Corporation, Law Dept. 2500 Renaissance Blvd, Suite 200 Gulph Mills, PA 19406</p> <p> SIGNATURE</p> <p>Aaron R. Ettelman NAME ATTORNEY FOR APPLICANT 42.516</p> <p>REGISTRATION NUMBER</p>			

"Express Mail" mailing label number EL541612377US.

PATENT  
Docket No. H 2938 PCT/US

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

RE: Serial No. 09/486,677  
PCT/EP98/05355  
International Filing Date: August 24, 1998  
Priority Date Claimed: September 1, 1997  
Applicant: Behler et al.  
Title: COLD-STABLE FATTY ALCOHOL ALKOXYLATES  
Applicants' Reference: H 2938 PCT/US

**PRELIMINARY AMENDMENT**

Commissioner for Patents  
Box PCT  
Washington, DC 20231

ATTN: DO/EO/US

Prior to the examination of the above-identified national stage application pursuant to the first submission under 35 U.S.C. §371 filed on March 1, 2000, please amend the English translation of the application submitted therewith, without prejudice, as follows:

**In the Specification:**

Please amend the instant Specification, without prejudice, as follows:

At page 1, please delete all text above line 7, and insert therefor the following:

**--TITLE OF THE INVENTION**

Random Fatty Alcohol Alkoxylates With Low Temperature Stability,  
and Methods of Producing and Using The Same

**BACKGROUND OF THE INVENTION--**

At page 2, between lines 28 and 29 thereof, insert:

**--BRIEF SUMMARY OF THE INVENTION--.**

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Serial No. 09/486,677**

At page 3, between lines 1 and 2, insert the following text:

--The present invention includes low-temperature stable random polymers of fatty alcohols with ethylene oxide and propylene oxide, the ethylene oxide and propylene oxide being present in a certain selected quantity ratio. The present invention also includes a process for the production of such random polymers of fatty alcohols, and to methods of using such polymers as surfactants in water-dilutable concentrates including, for example, concentrates of detergents, pesticides and/or agrochemicals.--

At page 3, between lines 19 and 20 thereof, insert:

**--DETAILED DESCRIPTION OF THE INVENTION--.**

At page 6, line 14 thereof, replace "2EO" with --2PO--.

At page 7, line 17 thereof, replace "(3.72 moles)" with --(3.77 moles)--.

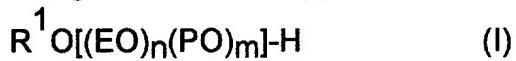
At page 8, between lines 1 and 2, insert --What is claimed is:--.

Please add new page 10, which is attached hereto, containing an Abstract of the Disclosure, following the claims.

**In the Claims:**

Please add new claims 10-30, as follow:

--10. (New) A mixture of polymers comprising random fatty alcohol alkoxylates according to the general formula (I):



wherein  $R^1$  represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents  $-CH_2CH_2O-$ , each PO independently represents  $-C(CH_3)HCH_2O-$  or  $-CH_2C(CH_3)HO-$ , and wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from

about 2 to about 7, and wherein m represents the average number of PO units present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3.--

--11. (New) The mixture according to claim 10, wherein n has a value of from about 3 to about 5.--

--12. (New) The mixture according to claim 10, wherein m has a value of from about 2 to about 2.5.--

--13. (New) The mixture according to claim 10, wherein n has a value of from about 3 to about 5, and m has a value of from about 2 to about 2.5.--

--14. (New) The mixture according to claim 10, wherein  $R^1O$  represents a fatty alcohol residue derived from a fatty alcohol mixture, said mixture comprising at least about 30% by weight of alcohols having from about 14 to about 18 carbon atoms and up to about 70% by weight of alcohols having from about 6 to about 12 carbon atoms.--

--15. (New) A process for producing a mixture of polymers comprising random fatty alcohol alkoxylates according to the general formula (I) :



wherein  $R^1$  represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents  $-CH_2CH_2O-$ , each PO independently represents  $-C(CH_3)HCH_2O-$  or  $-CH_2C(CH_3)HO-$ , and wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from about 2 to about 7, and wherein m represents the average number of PO units present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3; said process comprising reacting ethylene oxide, propylene oxide and fatty alcohol in the presence of an aqueous base, wherein the propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 10:90 to about 60:40.--

--16. (New) The process according to claim 15, wherein the

propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 10:90 to about 50:50.--

--17. (New) The process according to claim 15, wherein the propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 25:75 to about 50:50.--

--18. (New) The process according to claim 15, wherein the propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 25:75 to about 40:60.--

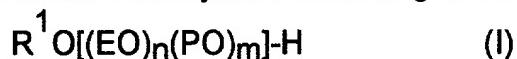
--19. (New) The process according to claim 15, wherein said fatty alcohol is a mixture of at least two fatty alcohols, said mixture comprising at least about 30% by weight of alcohols having from about 14 to about 18 carbon atoms and up to about 70% by weight of alcohols having from about 6 to about 12 carbon atoms.--

--20. (New) The process according to claim 15, wherein said aqueous base comprises a hydroxide selected from the group consisting of alkali metal hydroxides and alkali earth metal hydroxides.--

--21. (New) The process according to claim 20, wherein said hydroxide comprises potassium hydroxide.--

--22. (New) The product of the process according to claim 15.--

--23. (New) A water-dilutable concentrate comprising an active ingredient and a surfactant, said surfactant comprising a mixture of polymers comprising random fatty alcohol alkoxylates according to the general formula (I):



wherein  $R^1$  represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents  $-CH_2CH_2O-$ , each PO independently represents  $-C(CH_3)HCH_2O-$  or  $-CH_2C(CH_3)HO-$ , wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from about 2 to about 7, and wherein m represents the average number of PO units

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Serial No. 09/486,677**

present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3.--

--24. (New) The water-dilutable concentrate according to claim 23, wherein said active ingredient comprises one or more components selected from the group consisting of detergents, agrochemicals and pesticides.--

--25. (New) The water-dilutable concentrate according to claim 23, wherein said active ingredient comprises a detergent, and wherein said surfactant is present in an amount of from about 10% to about 30% by weight, based on the active ingredient.--

--26. (New) The water-dilutable concentrate according to claim 23, wherein said active ingredient comprises an agrochemical and/or a pesticide, and wherein said surfactant is present in an amount of from about 0.1% to about 15% by weight, based on the active ingredient.--

--27. (New) The water-dilutable concentrate according to claim 23, wherein n has a value of from about 3 to about 5.--

--28. (New) The water-dilutable concentrate according to claim 23, wherein m has a value of from about 2 to about 2.5.--

--29. (New) The water-dilutable concentrate according to claim 23, wherein n has a value of from about 3 to about 5, and m has a value of from about 2 to about 2.5.--

--30. (New) The water-dilutable concentrate according to claim 23,  
<sup>1</sup> wherein R<sup>1</sup>O represents a fatty alcohol residue derived from a fatty alcohol mixture, said mixture comprising at least about 30% by weight of alcohols having from about 14 to about 18 carbon atoms and up to about 70% by weight of alcohols having from about 6 to about 12 carbon atoms.--

Please cancel claims 1-9, without prejudice.

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**REMARKS**

Claims 10-30 are currently pending in the instant application.

The Specification has been amended to include the preferred section headings pursuant to 37 C.F.R. §1.77. The Specification has also been amended to correct two obvious typographical errors at pages 6 and 7. An Abstract of the Disclosure in accordance with the abstract of the corresponding international publication has been added on a separate sheet following the claims. It is submitted that the amendments to the Specification made herein introduce no new matter.

Their entry is therefore proper and respectfully requested.

Original claims 1-9 have been canceled and replaced with new claims 10-30 in an effort to place the claims in more proper U.S. format, with increased clarity, for the purposes of particularly pointing out and distinctly claiming that which Applicants regard as their invention. New claims 10-30 are supported by the claims as originally filed and in the Specification, for example, at page 3, lines 2-19, at page 4, lines 4-8, and 24-29, at page 5, lines 13-15, and at page 5, line 24 through page 6, line 11. No new matter has been introduced. Entry is therefore proper and respectfully requested.

**Case H 2938 PCT/US  
Serial No. 09/486,677**

Prompt examination of the instant application in view of the amendments made herein is respectfully requested.

Respectfully submitted,

**ANSGAR BEHLER, et al.**

June 26, 2000  
(Date)

  
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PATENT  
Docket No. H 2938 PCT/US

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

RE: Serial No. 09/486,677  
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International Filing Date: August 24, 1998  
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**PRELIMINARY AMENDMENT**

Commissioner for Patents  
Box PCT  
Washington, DC 20231

ATTN: DO/EO/US

Prior to the examination of the above-identified national stage application pursuant to the first submission under 35 U.S.C. §371 filed on March 1, 2000, please amend the English translation of the application submitted therewith, without prejudice, as follows:

**In the Specification:**

Please amend the instant Specification, without prejudice, as follows:  
At page 1, please delete all text above line 7, and insert therefor the following:

--TITLE OF THE INVENTION

Random Fatty Alcohol Alkoxylates With Low Temperature Stability,  
and Methods of Producing and Using The Same

**BACKGROUND OF THE INVENTION--**

At page 2, between lines 28 and 29 thereof, insert:

--BRIEF SUMMARY OF THE INVENTION--.

## Cold-stable Fatty Alcohol Alkoxylates

This invention relates to low-temperature-stable random polymers of fatty alcohols with ethylene oxide and propylene oxide, the ethylene oxide and propylene oxide being present in a certain selected quantity ratio, to a process for the production of such compounds and to the use of such 5 compounds as surfactants in water-dilutable concentrates, more particularly in concentrates of detergents, pesticides and agrochemicals.

Alcohol alkoxylates, which are also known as alkyl polyglycol ethers, are compounds which have been known for decades and which are obtained by reacting alcohols with ethylene oxide and/or propylene oxide.

- 10 The reaction takes place at elevated temperatures and pressures in the presence of acidic or alkaline catalysts. The use of basic compounds of the alkali and alkaline earth metals is of particular practical significance for the alkoxylation of fatty alcohols. Suitable basic compounds are alkali metal and alkaline earth metal alkoxylates, such as sodium methylate and 15 potassium methylate, or alkali metal and alkaline earth metal hydroxides, such as sodium hydroxide and potassium hydroxide. Recently, alkali metal and alkaline earth metal hydroxides have preferably been used as basic catalysts because, where alkali metal and alkaline earth metal alkoxylates are used, organic solvents, such as methanol, are essential. By contrast, 20 where the alkali metal and alkaline earth metal hydroxides are used, the reaction gives good yields, even in aqueous medium.

However, where aqueous solutions of alkali metal and alkaline earth metal hydroxides are used as basic catalysts, products with unfavorable low-temperature behavior which often tend to precipitate, even at room 25 temperature, are obtained, particularly in the ethoxylation of the fatty alcohols. This is attributable inter alia to the fact that the presence of

aqueous solutions of alkali metal and alkaline earth metal hydroxides promotes the formation of high molecular weight polyethylene glycols as secondary products. Although, in principle, these polyethylene glycols can be removed by extraction with suitable solvents, such as water, this does  
5 involve another process step which is very time-consuming and, in addition, cannot be universally applied.

According to the abstract of Japanese patent application 07,303,825 from the Journal CA Selects in: Alkoxylated Oleochemicals, Issue 1996, page 5, published by the American Chemical Society,  
10 Columbus, Ohio, No. 124, random adducts of C<sub>8-18</sub> alcohols containing 5 to 15 moles of ethylene oxide and 0.3 to 5.0 moles of propylene oxide show improved flow behavior at low temperatures. In the only example cited, lauryl alcohol is reacted with about 9 moles of ethylene oxide and about 2.4 moles of propylene oxide in the presence of potassium hydroxide as basic  
15 catalyst, a product with a pour point of 7.5°C being obtained. However, this product also tends to precipitate in storage at temperatures below 0°C which is problematical.

The problem addressed by the present invention was to provide fatty alcohol alkoxylates which would show very favorable low-temperature behavior and would not have any tendency to precipitate. The low-temperature behavior would be so pronounced that the products would remain clear liquids even at temperatures below 0°C so that no precipitation would occur, even in the event of storage at low temperatures.  
20 In addition, the products obtained would be readily soluble in cold water so that they would be suitable for use as surface-active compounds, for example in liquid concentrates of detergents. In addition, the compounds would lend themselves to production in the presence of aqueous solutions of bases.  
25

The problem stated above has surprisingly been solved by random polymers of fatty alcohols which contain ethylene oxide and propylene  
30

oxide in random-polymerized form in a certain selected ratio.

Accordingly, the present invention relates to random polymers of fatty alcohols with ethylene oxide and propylene oxide corresponding to formula (I):

5



in which  $R^1$  is an alkyl group containing 6 to 22 carbon atoms, EO stands CH<sub>2</sub>CH<sub>2</sub>O, PO stands for CHCH<sub>3</sub>CH<sub>2</sub>O and/or CH<sub>2</sub>CHCH<sub>3</sub>O, n is a whole or broken number of 2 to 7 and m is a whole or broken number of 1.5 to 3, characterized in that the molar ratio of propylene oxide to ethylene oxide is from 10:90 to 50:50.

Random polymers of fatty alcohols corresponding to formula (I), in which the molar ratio of propylene oxide to ethylene oxide is from 25:75 to 15 40:60, are particularly preferred.

In addition, random polymers of fatty alcohols corresponding to formula (I), in which n is a whole or broken number of 3 to 5, and random polymers of formula (I), in which m is a whole or broken number of 2 to 2.5, are preferred.

20 R<sup>1</sup> in formula (I) is derived from fatty alcohols with the formula R<sup>1</sup>OH which contain 6 to 22 carbon atoms. Fatty alcohols in this context are understood to be primary aliphatic alcohols in which R<sup>1</sup> is an aliphatic, linear or branched hydrocarbon radical containing 6 to 22 carbon atoms and 0 and/or 1, 2 or 3 double bonds. Typical examples are caproic alcohol, 25 caprylic alcohol, 2-ethylhexyl alcohol, capric alcohol, lauryl alcohol, isostearyl alcohol, myristyl alcohol, cetyl alcohol, palmitoleyl alcohol, stearyl alcohol, isostearyl alcohol, oleyl alcohol, elaidyl alcohol, petroselinyl alcohol, linolyl alcohol, linolenyl alcohol, elaeostearyl alcohol, arachyl alcohol, gadoleyl alcohol, behenyl alcohol, erucyl alcohol and brassidyl 30 alcohol and the technical mixtures thereof obtained, for example, in the

high-pressure hydrogenation of technical methyl esters based on fats and oils or aldehydes from Roelen's oxosynthesis and as monomer fraction in the dimerization of unsaturated fatty alcohols.

According to the invention, technical fatty alcohols containing at least 30% by weight C<sub>14-18</sub> fatty alcohols and at most 70% by weight C<sub>6-12</sub> fatty alcohols are preferred. The percentages by weight are based on the fatty alcohol mixture. Examples of such fatty alcohol mixtures are coconut, palm, palm kernel or tallow fatty alcohol.

The random polymers according to the invention are distinguished by very good low-temperature behavior, i.e. they are free-flowing and preferably clear even at temperatures below 0°C. The low-temperature behavior is determined by measurement of the cold cloud point in accordance with DIN ISO 3015. The products according to the invention are also distinguished by very good solubility in cold water, i.e. they have very short dissolving times. Dissolving behavior was determined by measuring the time which 10 g of the compound takes to form an optically clear solution in 90 g of stirred deionized water (temperature 23°C).

The present invention also relates to a process for the production of random polymers of fatty alcohols corresponding to formula (I) by reacting ethylene oxide and propylene oxide with fatty alcohols corresponding to the formula R<sup>1</sup>OH in the presence of aqueous bases, characterized in that propylene oxide and ethylene oxide in a molar ratio of 10:90 to 40:60 are reacted with fatty alcohols in known manner.

The random polymerization is a process known per se in which the above-described alcohols R<sup>1</sup>OH are reacted with propylene and ethylene oxide in a pressure vessel at temperatures of 120 to 190°C under a pressure of 3 to 5 bar. According to the invention, aqueous solutions of alkali metal and/or alkaline earth metal hydroxides, for example potassium hydroxide, are used as the basic compounds. The hydroxides are normally used in quantities of 0.2 to 5% by weight and preferably in quantities of 0.3

to 1.5% by weight, expressed as hydroxide and based on the mixture as a whole. It is advisable to use the hydroxides in the form of 40 to 60% by weight aqueous solutions. Propylene and ethylene oxide are reacted together with the fatty alcohols. To this end, they may be separately introduced into the reaction vessel through two different nozzles or may even be premixed in a mixing vessel and then introduced into the reactor. It is crucial to the invention that propylene oxide and ethylene oxide are used in the quantities and mixing ratios mentioned above. In general, ethylene oxide is used in a quantity of 2 to 7 moles and preferably 3 to 5 moles per mole fatty alcohol while propylene oxide is used in a quantity of 1.5 to 3 moles and preferably 2 to 2.5 moles per mole of fatty alcohol. A molar mixing ratio of propylene oxide to ethylene oxide of preferably 25:75 to 40:60 is of particular significance. The reaction of ethylene oxide and propylene oxide together with the fatty alcohols gives so-called random polymers, i.e. the addition takes place in statistical distribution.

If desired, the process according to the invention may be followed by neutralization of the basic catalyst. The neutralization step may be carried out with inorganic and/or organic acids, such as lactic acid, oxalic acid, citric acid, acetic acid, phosphoric acid or methane sulfonic acid. Neutralization to values of 6.5 to 7.5 is generally advisable when unsaturated fatty alcohols are used because they easily discolor under the influence of atmospheric oxygen. The products are obtained in the form of a 100% clear liquid.

The present invention also relates to the use of the above-described random polymers of fatty alcohols as surface-active agents in water-dilutable concentrates, for example in concentrates of detergents or in concentrates of pesticides and agrochemicals. In recent years, detergents, for example, have been increasingly marketed as concentrates which contain only a little water. Such concentrates can be diluted with water before use by the end user. The concentrates are expected to dissolve

quickly without forming any precipitates. This is achieved by using the compounds according to the invention as surface-active agent. The products according to the invention may be used in detergent concentrates in quantities of 10 to 30% by weight, based on active ingredient in the 5 concentrate. The concentrates may additionally contain typical constituents, such as anionic surfactants, nonionic surfactants, preservatives, solubilizers, etc., which are typical of detergents, in the usual quantities.

The products according to the invention may be used in pesticide and agrochemical concentrates in quantities of 0.1 to 15% by weight, 10 based on active principle in the concentrate. In this case, too, the concentrates may contain typical constituents in typical quantities.

### Examples

#### 1. Production of a random polymer of a C<sub>12/14</sub> alcohol+5EO+2EO

15 366.3 g (1.89 moles) of a C<sub>12/C<sub>14</sub></sub> fatty alcohol mixture (about 40% C<sub>12</sub> and 60% by weight C<sub>14</sub>) were introduced into a pressure vessel with 5 g of a 50% by weight aqueous potassium hydroxide solution. The vessel was then evacuated for 30 minutes at 100°C and subsequently purged with nitrogen. A mixture of 414.9 g (9.43 moles) of ethylene oxide and 218.8 g 20 (3.77 moles) of propylene oxide was then introduced at 120°C. The maximum pressure was 5 bar. On completion of the reaction, the mixture was stirred for 1 hour at 120°C and the reactor evacuated for another 30 minutes at 120°C. The product obtained was neutralized with lactic acid.

A clear liquid product with a cold cloud point of -4°C was obtained; 25 the dissolving time in water was 5 seconds.

#### 2. Production of a random polymer of a C<sub>12/14</sub> alcohol+3EO+2PO

As in Example 1, 439.1 g (2.26 moles) of a C<sub>12/C<sub>14</sub></sub> fatty alcohol were reacted with a mixture of 298.5 g (6.78 moles) of ethylene oxide and 30 262.4 g (4.52 moles) of propylene oxide in the presence of 5 g of a 50% by

weight aqueous potassium hydroxide solution.

A clear liquid product with a cold cloud point of -14°C which took 3 seconds to dissolve in water was obtained.

5 Comparison Example 1: Block polymer of a C<sub>12/14</sub> alcohol with 1PO+5EO+1PO

In accordance with Example 1, the quantities of fatty alcohol described in Example 1 were reacted first at 120°C with 109.4 g (1.89 moles) of propylene oxide, then at 180°C with 414.9 g (9.43 moles) of ethylene oxide and, on completion of the reaction, at 120°C with another 10 109.4 g (1.89 moles) of propylene oxide.

A clear liquid product with a cold cloud point of 1.5°C was obtained. The dissolving time in water was 75 seconds.

Comparison Example 2: Block polymer of a C<sub>12/14</sub> alcohol with 5EO+2PO

15 In accordance with Example 1, the quantity of fatty alcohol described in Example 1 was reacted first at 180°C with 414.9 g (9.43 moles) of ethylene oxide and then at 120°C with 218.8 g (3.72 moles) of propylene oxide.

A clear liquid product with a cold cloud point of 7.5°C was obtained. 20 The dissolving time in water was 8 seconds.

## **CLAIMS**

1. Random polymers of fatty alcohols with ethylene oxide and propylene oxide corresponding to formula (I):

$$5 \quad R^1O(EO)_n(PO)_mH \quad (I)$$

in which R<sup>1</sup> is an alkyl group containing 6 to 22 carbon atoms, EO stands for CH<sub>2</sub>CH<sub>2</sub>O, PO stands for CHCH<sub>3</sub>CH<sub>2</sub>O and/or CH<sub>2</sub>CHCH<sub>3</sub>O, n is a whole or broken number of 2 to 7 and m is a whole or broken number of 1.5 to 3,

10 characterized in that the molar ratio of propylene oxide to ethylene oxide is from 10:90 to 50:50.

2. Random polymers as claimed in claim 1, characterized in that the molar ratio of propylene oxide to ethylene oxide is from 25:75 to 40:60.

3. Random polymers as claimed in claim 1, characterized in that n is a whole or broken number of 3 to 5.

4. Random polymers as claimed in claim 1, characterized in that m is a whole or broken number of 2 to 2.5.

5. Random polymers as claimed in claim 1, characterized in that R<sup>1</sup> is derived from a fatty alcohol mixture containing at least 30% by weight of

20 C<sub>14-18</sub> fatty alcohols and at most 70% by weight of C<sub>6-12</sub> fatty alcohols.

6. A process for the production of random polymers of fatty alcohols with ethylene oxide and propylene oxide corresponding to formula (I):

$$R^1O(EO)_n(PO)_mH \quad (I)$$

25

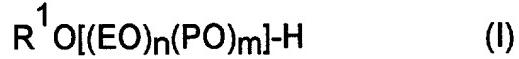
with the definitions given in claim 1, by reacting ethylene oxide and propylene oxide with fatty alcohol having the formula  $R^1OH$  in the presence of aqueous bases, characterized in that the propylene oxide and ethylene oxide in a molar ratio of 10:90 to 40:60 are reacted with fatty alcohols by

30 methods known per se.

7. A process as claimed in claim 6, characterized in that the molar ratio of propylene oxide to ethylene oxide is in the range from 25:75 to 40:60.
  8. A process as claimed in claim 6, characterized in that a fatty alcohol mixture containing at least 30% by weight of C<sub>14-18</sub> fatty alcohols and at 5 most 70% by weight of C<sub>6-12</sub> fatty alcohols is reacted.
  9. The use of the random polymers of fatty alcohols corresponding to formula (I) claimed in claim 1 as a surfactant in water-dilutable concentrates.

## ABSTRACT OF THE DISCLOSURE

Random fatty alcohol alkoxylates with low temperature stability which are polymerization products of fatty alcohols, ethylene oxide and propylene oxide, according to the general formula (I) :



wherein  $R^1$  represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents  $-CH_2CH_2O-$ , each PO independently represents  $-C(CH_3)HCH_2O-$  or  $-CH_2C(CH_3)HO-$ , wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from about 2 to about 7, and wherein m represents the average number of PO units present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3; are disclosed. A process for the production of such random fatty alcohol alkoxylates and methods of their use in water-dilutable concentrates are also disclosed.

Type a plus sign (+) inside this box → 

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0010/PTO Rev. 6/95	U.S. Department of Commerce Patent and Trademark Office	Attorney Docket Number	H 2938 PCT/US
<b>DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION</b>		First Named Inventor	BEHLER, Ansgar
<b>COMPLETE IF KNOWN</b>			
		Application Number	09/486,677
		Filing Date	06/26/2000
		Group Art Unit	
		Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**COLD-STABLE FATTY ALCOHOL ALKOXYLATES**

(*Title of the Invention*)

the specification of which

is attached hereto

OR

was filed on (MM/DD/YYYY)  as United States Application Number or PCT International

Application Number  and was amended on (MM/DD/YYYY)  (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES	Certified Copy Attached? NO
197 38 108.1	Germany	09/01/1997	<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>	<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>	<input checked="" type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>

Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

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**DECLARATION****Page 2**

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U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP98/05355	08/24/1998	

Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

Firm Name \_\_\_\_\_ Customer Number or label \_\_\_\_\_  
 OR  
 List Attorney(s) and/or agent(s) name and registration number below:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:		<input type="checkbox"/> A petition has been filed for this							
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Inventor's Signature	<u>Ansgar Behler</u>					Date	<u>03/23/2000</u>		
Residence: City	Bottrop DEX	State		Country	Germany	Citizenship	Germany		
Post Office Address	<u>Siegfriedstrasse 80</u>								
Post Office Address									
City	46240 Bottrop	State		Zip		Country	Germany	Applicant Authority	
<input checked="" type="checkbox"/> Additional inventors are being named on supplemental sheet(s) attached hereto									

**DECLARATION****ADDITIONAL INVENTOR(S)**  
Supplemental Sheet

Name of Additional Joint Inventor, if any:  A petition has been filed for this unsigned inventor

2-00

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City	40699 Erkrath	State	Zip	Country	Germany	Applicant Authority		
Name of Additional Joint Inventor, if any:		<input type="checkbox"/> A petition has been filed for this unsigned inventor						
Given Name			Middle Initial	Family Name				Suffix e.g. Jr.
Inventor's Signature						Date		
Residence: City			State	Country			Citizenship	
Post Office Address								
Post Office Address								
City	State	Zip	Country				Applicant Authority	
Name of Additional Joint Inventor, if any:		<input type="checkbox"/> A petition has been filed for this unsigned inventor						
Given Name			Middle Initial	Family Name				Suffix e.g. Jr.
Inventor's Signature						Date		
Residence: City			State	Country			Citizenship	
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